Microbial Methane Oxidation at the Fíflholt landfill in Iceland

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Methane oxidizing biosystems have received wide recognition in recent years as a cost effective and important means to reduce emissions from landfills. However, there is no documentation of the oxidation capacity of Icelandic landfill covers to date and there is limited information on microbial methane oxidation in boreal climates. The present study was carried out to qualitatively assess the oxidation capacity of the current top cover of one of the cells of the Fíflholt landfill, located in West Iceland, using the gas profile method (CO_2/CH_4 ratio). The landfill has no gas recovery system and the cover is composed of 15-25 cm crushed wood overlain by about 1 m of gravelly sand with 7% (w/w) organic matter content. Sampling probes were installed at two locations on cell 2 at different depths (5 to 120 cm). Several gas concentration measurements were carried out during the autumn and the winter of 2012-2013. It was observed that atmospheric air penetrated deep into the cover and oxidation activity was observed in the gas profiles. The oxidation zone was situated mainly below 40 cm from the surface and went as deep as about 1 m below surface, i.e. to the base of the cover. Oxidation efficiencies ranged from 0 to 99%, reaching maximal values between 30 and 60 cm depth, with mean values 59% and 77% respectively for the two sampling locations for the whole study period. It must be highlighted that relatively high oxidation efficiencies were obtained during winter. This indicates that methane oxidation can occur yearlong in Iceland, although a more thorough field study is needed to verify the extent of this phenomena in Icelandic landfill covers. Such a study would also permit to verify the applicability of the default value of 10% methane oxidation used in biogas generation and emission models.

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Yfirborðslög sem innihalda lífræn efni og oxa metan hafa hlotið viðurkenningu á undanförnum árum sem hagkvæm og áhrifarík leið til að draga úr losun gróðurhúsalofttegunda frá urðunarstöðum. Oxunarhæfni yfirborðslaga á íslenskum urðunarstöðum hefur hinsvegar ekki verið rannsökuð og það er takmörkuð þekking á metanoxun örvera í köldu loftslagi. Þessari rannsókn var hleypt af stað til að meta hlutfallslega oxunarhæfni núverandi yfirborðslags í rein 2 á urðunarstaðnum í Fíflholtum á Mýrum með því að nota gasprófílaðferðina (hlutfall CO_2/CH_4). Urðunarstaðurinn er ekki búinn gassöfnunarkerfi og yfirborðið er samansett af 15-25 cm timburkurli undir u.þ.b. 1 m af malarkenndum sandi með 7% (w/w) lífrænu innihaldi. Mælirör voru sett niður á tveim stöðum á rein 2 á mismunandi dýpi (5 - 120 cm) og punktmælingar gerðar á haust- og vetrarmánuðum 2012-2013. Niðurstöður sýna að andrúmsloft smýgur almennt djúpt niður í yfirborðslagið og mikil oxunarvirkni sást í gasprófílum. Oxunin átti sér aðallega stað á 40 cm dýpi og neðar, mögulega alveg frá botni yfirborðslagsins eða frá 1 m dýpt. Oxunarhlutfall var á bilinu 0 til 99% og náði hámarki á 30 - 60 cm dýpi, eða meðalgildum 59% og 77% fyrir hvort tveggja staðanna á öllu tímabilinu. Athygli vakti að tiltölulega mikil oxun átti sér stað um vetur. Þetta bendir til þess að oxun metans geti átt sér stað árið um kring á Íslandi, en frekari vettvangsrannsókna er þörf til að sannreyna umfang þessa fyrirbæris í íslenskum urðunarstöðum. Slík rannsókn myndi einnig sannreyna hvort að sjálfgefið 10% oxunarhlutfall, sem notað er víða í líkönum sem meta myndun og losun hauggass, eigi við.

1. Introduction

Landfill gas is produced through microbial anaerobic degradation of organic waste and is mainly composed of methane and carbon dioxide, typically in the concentrations of 55-60% methane and 40-45 % carbon dioxide (Scheutz et al., 2009). Methane is a greenhouse gas up to 25 times more potent than carbon dioxide over a 100-year period, as it is more effective at absorbing infrared radiation (IPCC, 2007). Landfills are estimated to contribute with up to 5% of overall global greenhouse gas emissions today or about 18% of global CH_4 emissions (Bogner et al., 2007). They rank among the largest anthropogenic CH_4 sources worldwide - second largest in Europe (EEA, 2009) - making them a good target for mitigation (e.g. Forster et al., 2007).

Methane emissions from landfills is a product of landfill gas generation, gas recovery and microbial CH_4, which largely depends on both site-specific soil characteristics and meteorological factors. Many modern landfills have gas collection systems that either extract the gas for energy production or incineration. With the adoption of the EU Landfill directive through Icelandic regulation no. 738/2003 on landfill waste, landfills in Iceland receiving biodegradable waste were required to collect landfill gas for utilization or flaring after July 16, 2009.

Gas collection systems can only recover a fraction of the gas due to leaks in the system and because of fugitive gas emissions that escape through cracks or other preferential pathways in the landfill cover (e.g. Christophersen & Kjeldsen, 2011; Börjesson et al., 2007; Scheutz et al., 2009; Scheutz et al., 2011). In smaller and older landfills, methane production is too low for recovery or flaring, and installation of a gas extraction system is inefficient (Huber-Humer et al., 2009), thus allowing all of the generated gas to pass through the cover soil. In shallow landfills (<8 m depth), which are common in Iceland, the installation of gas extraction systems is furthermore technically either very difficult or near impossible (Scharff et al., 2011), whether it be with horizontal or vertical wells. Such a system is also likely to partially take in atmospheric air, highly reducing the recovery efficiency, as all Icelandic landfills to date do not have a top liner.

The technique of enhancing the activity of methanotrophs in landfill cover soils to oxidize methane has received wide recognition in past years as a cost-effective and important means to reduce fugitive emissions (e.g. Gebert and Gröngröft, 2006; Huber-Humer et al., 2009; Scheutz et al., 2009; Cabral et al., 2010; Chanton et al, 2011, Roncato & Cabral., 2012). It serves as a complementary strategy for methane emissions escaping gas collection, and for emissions mitigation at smaller and older sites without gas recovery systems. Furthermore, since gas is still being generated after landfills are no longer in operation, landfill after-care with oxidizing biosystems is considered among key mitigating measures to reduce long-term greenhouse gas emissions from landfills (Bogner et al., 2007).

Metanotrophs are a certain class of prokaryotic bacteria that consume methane as their only source of carbon and energy. At lower flow rates, methanotrophic bacteria can consume a larger portion of the methane delivered. A passive methane oxidizing system can therefore be considered a viable management approach for the treatment of fugitive emissions at landfills with recovery systems, or for landfills with a low methane generation rate per area due to size or age of wastes.

The majority of Icelandic landfills receiving biodegradable waste are relatively small. Aside from the three largest landfills currently in operation (Álfsnes, Stekkjarvík, Fíflholt), all other landfills in Iceland receive <5,000 tons of waste per year, and the waste degradation rate and thus methane generation rate in small Icelandic landfills is likely lower than in landfills in a moderate European climate (Scharff et al., 2011, Kamsma & Meyles, 2003). A study carried out in 2010 (Júlíusson, 2011) furthermore suggested that most landfills in Iceland generate too little methane for it to be technically or economically feasible to collect biogas, as required by regulation.

No documentation on the oxidation capacity of Icelandic landfill covers exists to date. Since there is limited information on microbial methane oxidation in boreal climates, a recent TAIEX mission report (Scharff et al., 2011) recommended a demonstration project to gather information on the current situation. The Icelandic Association of Local Authorities therefore launched a project in cooperation with The Solid Waste Management of West Iceland Regional Office, EFLA Consulting Engineers and the University of Iceland to study and assess the oxidation capacity of the current top cover in cell 2 at the Fíflholt landfill in West Iceland, 64°N, using the gas profile method (CO_2/CH_4 ratio) (Gebert et al., 2011b). In addition to assessing the oxidation efficiency of the cover, the method enables identification of the optimum zone for methane oxidation or oxidation horizon, i.e. the depth at which environmental conditions such as temperature and moisture promote methanotrophic growth.

The objective of this study was to investigate whether there was evidence of methane oxidation in the top cover at the Fíflholt landfill, particularly during colder months of the year, and to examine whether this oxidation could be assessed to a certain extent, i.e. using the gas profile method. This information will prove valuable in the near future when performing emissions estimations, for individual landfills in Iceland and for National Inventory Reports on greenhouse gas emissions.

2. Materials and Methods

2.1. Study site and measurements

The Fíflholt landfill currently receives up to 10,000 tpa of waste, including biodegradable waste from rural and urban areas in the region. It is situated roughly 10 km from the shore and has been in operation since 1999. It is one of the larger Icelandic landfills, although small in international comparison, and has no gas collection system. The gas generated in the landfill therefore migrates freely through the top cover. The top cover of the landfill is 1 - 1.2 m thick and is composed of 15 - 25 cm of shredded wood mulch overlain by approximately 1 m of excavated local soil.

Stainless steel sampling probes of different depths (5, 10, 20, 30, 40, 60, 80 and 120 cm) were installed at two locations at cell 2, about 100 m apart, and sealed with rubber stops (Figure 1). The two locations were identified as C2-N and C2-S, in proximity to two previously installed gas monitoring wells. Cell 2 is about 3-5 m deep, covers 0.9 ha of surface area, and received waste between the years 2003 and 2006, i.e. a total of 36,000 tons of waste. Monthly point measurements of gas composition (CH_4, CO_2, O_2, and N_2) at probe depth were carried out from August 2012 to February 2013, and again in August 2013. During each measurement, after initial purge, gas from the probes was extracted using a syringe and needle and fed into a portable gas analyser (Geotechnical Instruments GA2000 Plus). Soil temperatures at probe depths were also measured on selected dates, using a

FLUKE 54-II thermometer. More information on data procedures and results can be found in Kjeld (2013).

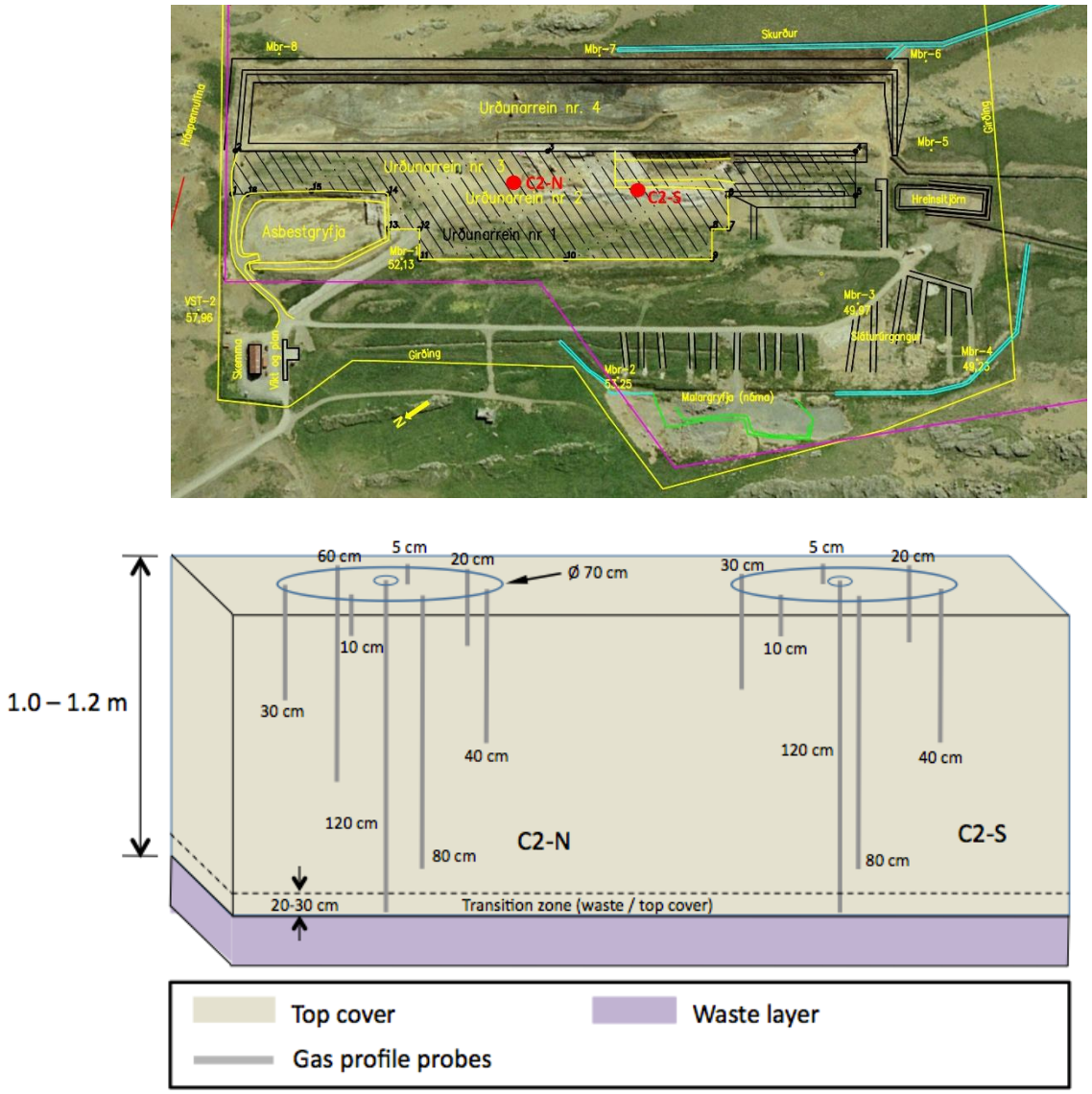

Figure 1 *Plan view (upper figure) and profile view (lower figure) of cell 2 of the Fíflholt landfill. The upper figure shows the location of C2-N and C2-S on cell 2 (VERKÍS Consulting Engineers) and the lower figure shows the gas probe setup at both locations.*

Meteorological data was collected from the on-site weather station run by the Iceland Meteorological Office. A soil sample from an approx. 1 m^3 excavation of the top cover in the middle of cell 2 was extracted in December 2012, divided into two subsamples, and analysed at the Innovation Center Iceland. As the top cover soil at Fíflholt is fairly homogeneous, one sample was regarded sufficient to roughly estimate organic matter content and grain size distribution. The soil analysis revealed a highly porous gravelly sand with 7% organic matter content (loss-on-ignition test).

2.2 Gas profile method (CO_2/CH_4 ratio) and oxidation efficiency (Eff_{ox}) calculation

The gas profile method is based on the change in the ratio of CO_2 to CH_4 in the gas profile, compared to the ratio in the raw landfill gas (Gebert et al, 2011b). It is assumed that the change occurs as a result of an oxidation process, i.e. the following reaction:

$$CH_4 + 2\ O_2 \rightarrow CO_2 + 2\ H_2O \quad (1)$$

The ratio CO_2 to CH_4 is higher near the surface, since CH_4 gradually gets converted to CO_2 in the oxidation process. The method is based on a few assumptions (Gebert et al., 2011b); e.g. that the size of the methanotrophic population is stable in the landfill cover, i.e. no net transfer of carbon into the microbial biomass; that the system is under steady state; and that the net increase in CO_2 is due to oxidation of CH_4 only, i.e. microbe respiration plays a minor role in the production of CO_2. The last assumption applies for biofilters with high CH_4 loading and oxidation rates, in daily and temporary landfill covers, and it is assumed that it can be applied for medium sized landfills without gas extractions systems, such as those found in Iceland.

The method assumes that the volume of CO_2 produced equals the volume of CH_4 oxidized, which can be derived from the following equation:

$$\frac{C_{2(Q\ \ )} + x}{C_{4(H\ \ )} - x} = {}_{G}\frac{C_{2(Q)}}{C_{4(H)}} \quad (2)$$

where x = share of oxidized CH_4 (%) at a certain depth i, $CH_{4(LFG)}$ = CH_4 concentration of the landfill gas (%), $CO_{2(LFG)}$ = CO_2 concentration of the landfill gas (%), $CH_{4(i)}$ = CH_4 concentration at depth i (%) and $CO_{2(i)}$ = CO_2 concentration at depth i (%). From the above assumptions it follows that $CO_{2(LFG)} = 100 - CH_{4(LFG)}$, i.e. other landfill gasses are in trace amounts (≤1%) and considered negligible.

For calculations, values of $CH_{4(LFG)}$ and $CO_{2(LFG)}$ were applied where $CH_{4(LFG)}$ concentrations were the highest, sometimes from monitoring wells and sometimes from the deepest sampling probes. The oxidation efficiency, Eff_{ox}, is obtained by dividing the share of oxidized CH_4 at each monitored depth, x, by the concentration of CH_4 in the landfill gas, $CH_{4(LFG)}$, i.e.

$$E\ f_{o}\ \not{f}_{x} \frac{x}{C\ H_{4(L\ \ F\ \ G}} \quad (3)$$

In the soil profile, this efficiency represents the cumulative percentage of CH_4 oxidized. The higher the efficiency, the more methane oxidized. The method is independent of the nature of the flux (diffusive or advective) of both landfill gas seeping up through the cover and of the influx of atmospheric gas from the surface. The method also assumes that CH_4 and CO_2 are diluted to the same extent in the pore volume by atmospheric gases.

3. Results and discussion

3.1 Gas profiles

A total of 8 gas concentration profiles were obtained at C2-N and 6 profiles at C2-S. Figure 2 shows two typical gas concentration profiles obtained during the study period,

i.e. one showing evidence of high oxidation (Figure 2a) and little or no oxidation (Figure 2b). A high oxidation profile was the most common profile during the study.

The concentration of nitrogen, N_2, can be used as a tracer gas, since N_2 is neither produced nor consumed in the cover. At both locations, it was observed that atmospheric gases O_2 and N_2 were in significant concentrations at 80 cm depth (32-73%). This indicates that atmospheric air penetrates deep into the soil cover, creating part of the conditions needed for methane oxidation to occur. This deep penetration of atmospheric air was expected, given the coarse texture of the soil cover.

An indication of methane oxidation can be inferred from gas concentration profiles, where the CH_4 and CO_2 profiles converge from the bottom to the top of the profile (Figure 2a), thus increasing the CO_2/CH_4 ratio (see Eq. 2). At C2-N, this phenomena was observed at 80 cm depth, indicating an existing oxidation horizon right from the bottom of the top cover. At C2-S, the increase in the CO_2/CH_4 ratio was observed between 40 and 80 cm depth, indicating that the oxidation horizon was situated slightly higher at that location.

On a few occasions, such as in December 2012 at both locations and in August 2013 at C2-N, the gas concentration profiles did not converge and thus little or no increase in the CO_2/CH_4 ratio occurred. This was coupled with little observed penetration of atmospheric air (Figure 2b), indicating that little or no oxidation took place during these sampling periods.

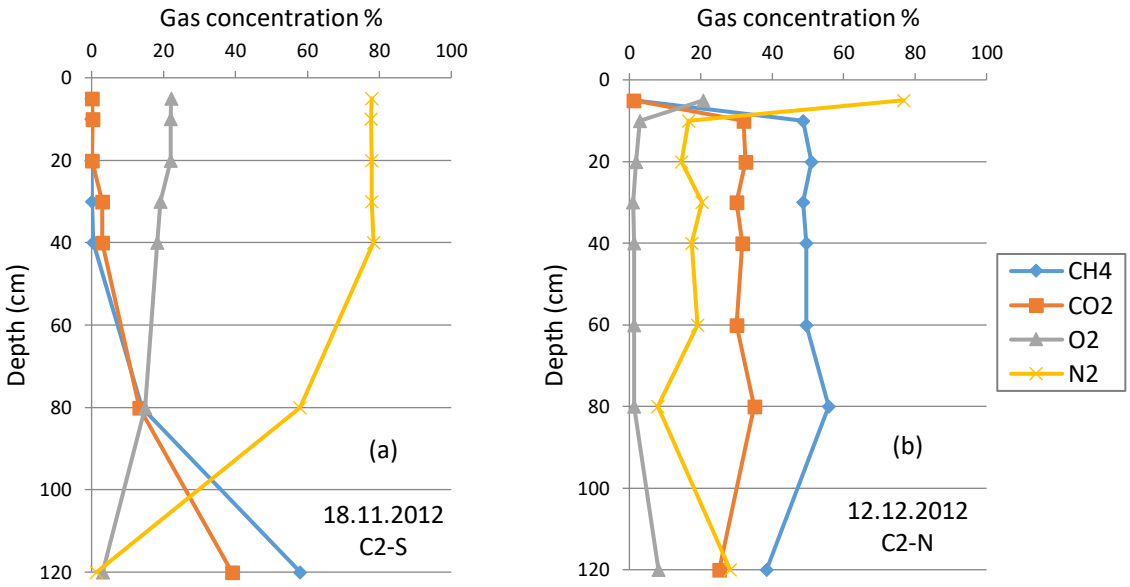

Figure 2 *Two exemplary soil gas profiles from the study: (a) C2-S on Nov 18, 2012; (b) C2-N on Dec 12, 2012.*

3.2 Methane oxidation efficiency, Eff_{ox}

Oxidation efficiencies within the soil profiles, calculated according to Eq. 3, are shown in Figures 3 and 4 for locations C2-S and C2-N, respectively. The average oxidation profiles for the entire study period are also shown. Oxidation efficiencies ranged from 0 to 99%. As can be observed, Eff_{ox} increased moving up the profile and reached a maximum value between 30 cm and 60 cm depth, a clear indication of the position of the oxidation horizon.

Average Eff_{ox} values for the entire study period attained a maximum of 59% at 60 cm for C2-N and 77% at 30 cm for C2-S. It must be emphasized that the oxidation efficiency is neither an accurate nor constant value, and that this method is only intended to give an indication of the overall efficiency (Gebert et al. 2011b).

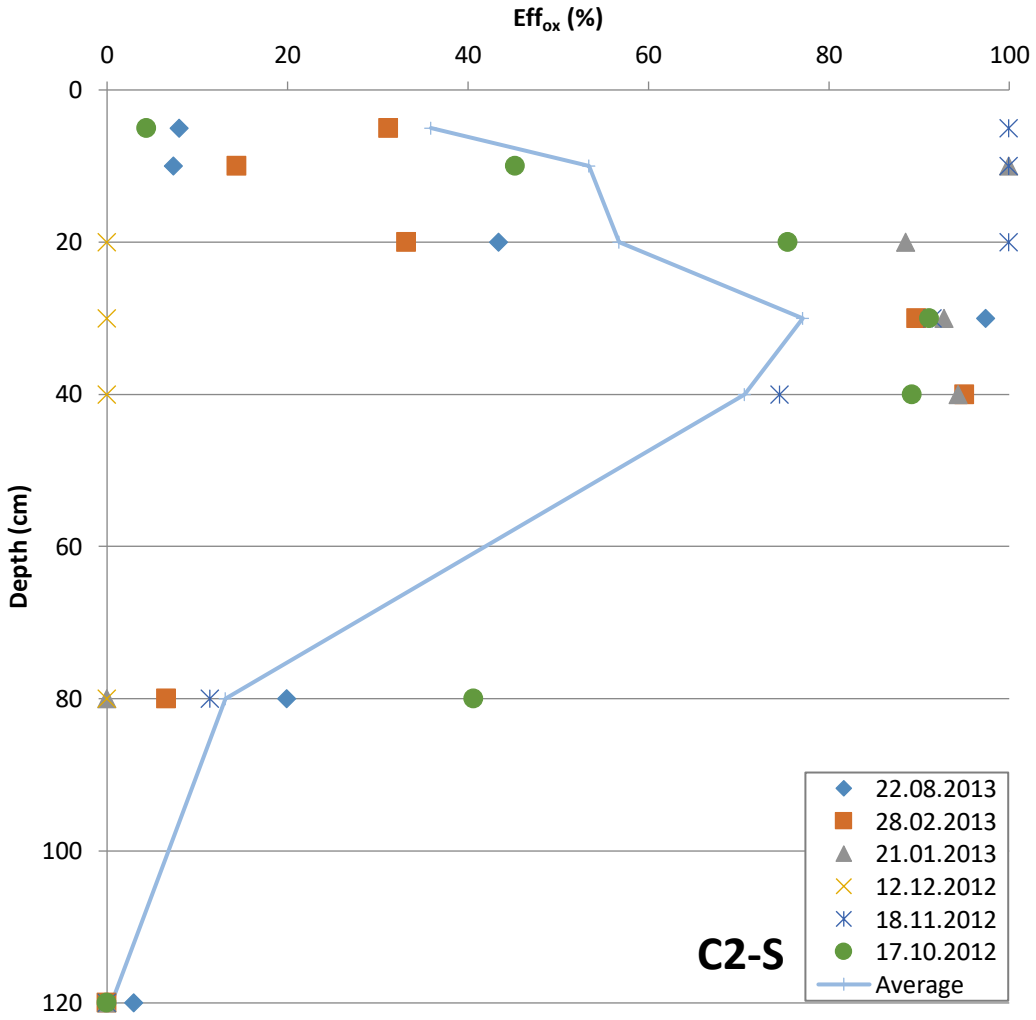

Figure 3 *Methane oxidation efficiency Eff_{ox} vs. depth during the study period at C2-S.*

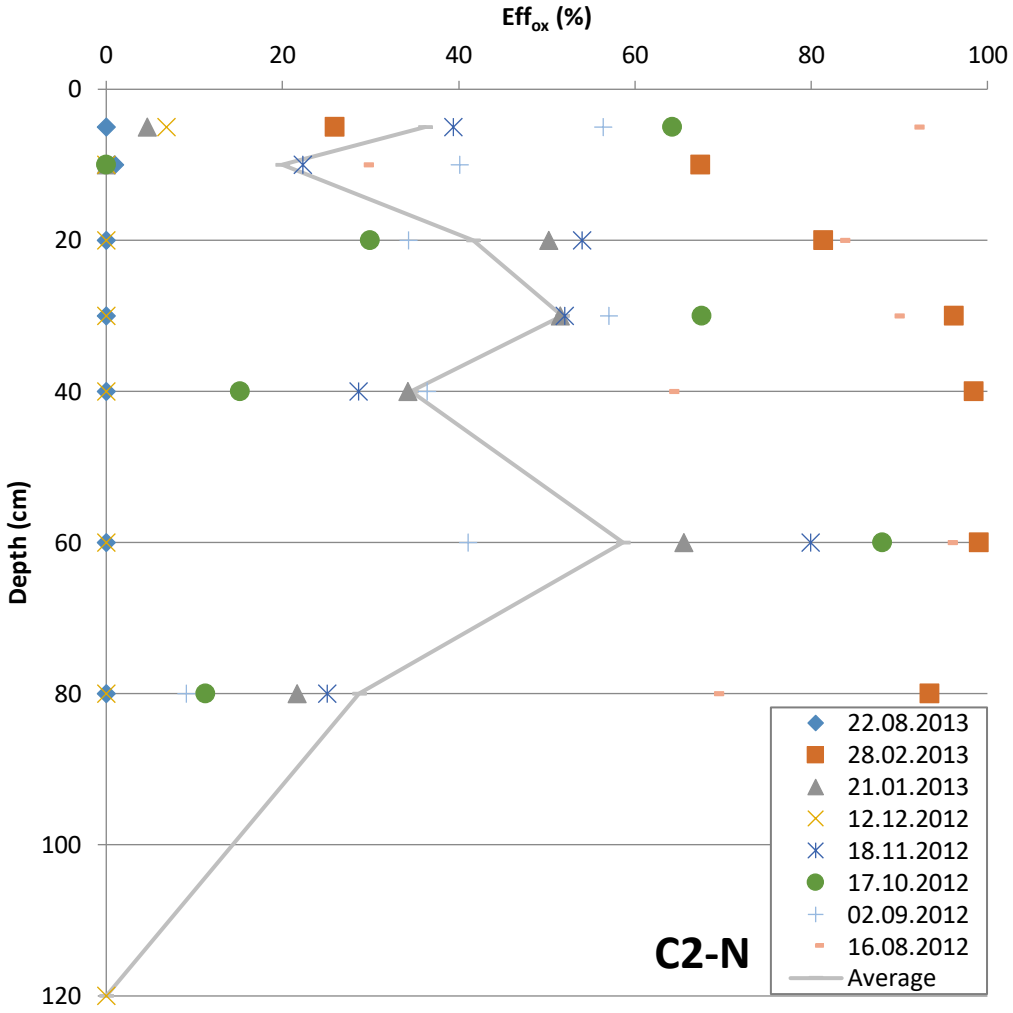

Figure 4 *Methane oxidation efficiency Eff_{ox} vs. depth during the study period at C2-N.*

A reduction in efficiency was commonly observed in the shallowest tubes at both locations, i.e. from depth 40 cm and upward. Since CH_4 and CO_2 concentrations were generally very low in the top 40 cm (typically <1% at C2-S and <15% at C2-N), the drop in efficiency may result from CO_2 dilution by atmospheric N_2 and O_2, present in concentrations quite close to those in the air. This tends to skew the CO_2/CH_4 ratio and therefore reduce the accuracy of the efficiency calculation.

3.3 Spatial variability

There were some differences observed between C2-N and C2-S, indicating spatial variability within cell 2. The intrusion of atmospheric air and depth of oxidation was slightly more pronounced at C2-N than at C2-S. There was also more consistency in the data collected from C2-S (Figure 3), whereas profile data from C2-N showed more disparities (Figures 4). During the time of measurement, the age of the waste at C2-N was about 9 years and about 7 years at C2-S. The waste composition changed slightly between years, with more biodegradable waste below C2-S. The differences observed between C2-N and C2-S do not conform with the age or the biodegradability of the waste body below, indicating that the observed variability between the two locations it is rather due to characteristics and conditions of the overlying top cover. Oxidation activity depends on soil texture, specifically on pore size distribution and on soil moisture retention capacity (e.g. Röwer et al., 2011). A higher share of air-filled pores

increases the availability for gaseous transport. During the study period, the 120-cm tube at C2-N was filled with water on every sampling date, except in December 2012, indicating that the bottom of the top cover was typically saturated at that location, except during very dry periods such as encountered in December. The presence of decomposing chopped wood at this depth may have caused accumulation of moisture and this might have driven landfill gases to migrate via preferential pathways such as cracks in the cover or via lateral diffusion to areas adjacent to the cell. Such phenomena could explain the greater variability in oxidation efficiencies with time at this location (Figure 4), and the generally lower oxidation efficiencies, as compared to C2-S.

3.4 Influence of meteorological parameters

In addition to soil moisture and texture, the microbial oxidation process is sensitive to climatic factors including temperature and barometric pressure changes (e.g. Gebert et al., 2011a; Börjesson et al., 2004), many of which are interrelated. In this study, soil temperatures at probe depths were measured from November to February, and also in August 2013. Figure 5 shows the relationship between calculated oxidation efficiencies and soil temperatures with depth for both sampling locations, C2-S and C2-N. It is quite striking to observe that high efficiencies were calculated despite the generally low temperatures (2-12 °C) at the bottom of the cover. Also worthwhile noting is the fact that the same level of oxidation efficiency was calculated for higher (~12 °C) and lower (2-4 °C) temperatures. Temperature has been observed to be one of the controlling factors in the oxidation process in a number of studies, and in warmer climates, optimum oxidation has been reported at much higher soil temperatures, i.e. around 25-35°C (Scheutz et al., 2009). Oxidation has, however, also been reported in colder areas at temperatures down to 2°C (Christophersen et al., 2000) and 1°C (Einola et al., 2007), although documentation is sparse for colder climates. The limited data base of this study seems to indicate that methane oxidation can take place throughout the year, and that the low temperatures prevailing yearlong in Iceland are not the most important factor controlling methane oxidation.

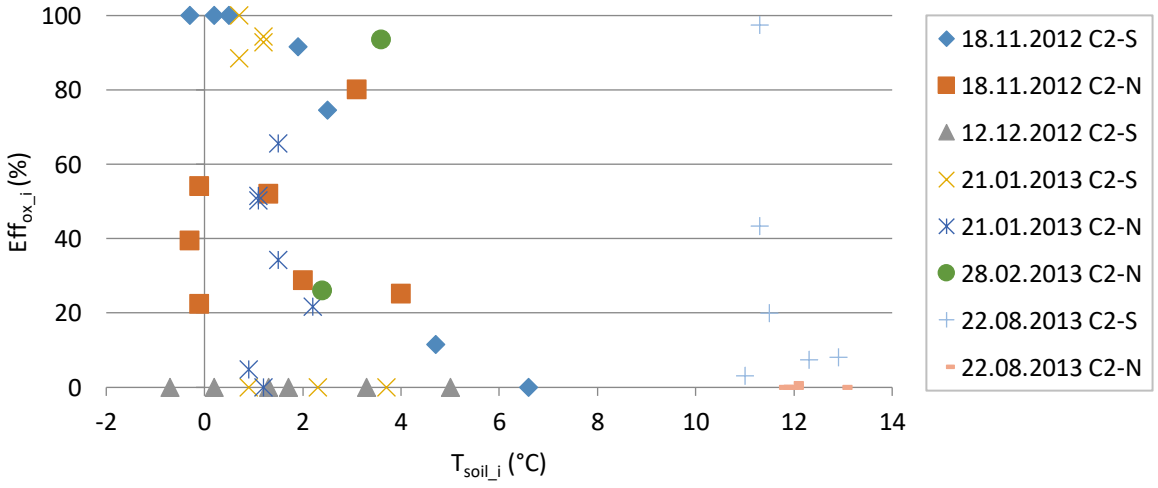

Figure 5 *Soil temperature at depth i, T_{soil_i} (°C), vs oxidation efficiency at depth i, Eff_{ox_i} (%), for both measurement locations C2-S and C2-N during the study period.*

A number of studies have shown a relationship between pressure change and landfill gas release through the top cover (e.g. Kjeldsen & Fischer, 1995; Gebert & Gröngröft, 2006). When atmospheric pressure drops rapidly, the formed pressure gradient can lead to advective gas transport out of the waste layer. This can result in higher emissions, possibly overriding

the maximum oxidation rate of the soil and resulting in lower efficiencies. Over the study period, atmospheric pressures between 1001 and 1031 hPa were recorded and it did not undergo any significant changes during sampling. During the study period, the most important pressure drops were observed in December 2012 and in August 2013, where over the course of 12 hours the pressure dropped by about 5 hPa or the equivalent of a 5-cm water column, see Figure 6. This is a relatively mild drop in atmospheric pressure and the decrease in oxidation efficiencies observed on those dates can therefore not be associated with changes in atmospheric pressure. Furthermore, the high efficiencies at C2-S vs zero efficiencies at C2-N in August 2013 indicate that other environmental factors than temperature or atmospheric pressure, e.g. water content, were more important in the oxidation process.

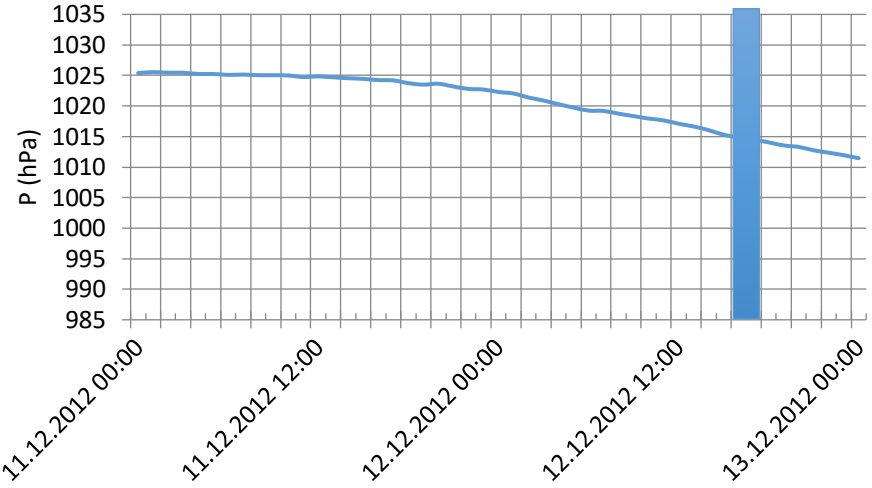

Figure 6 *48-hour atmospheric pressure (hPa) development prior to and after sampling on December 12, 2012. The blue vertical line denotes the time of sampling.*

Based on the sparse data on accumulated precipitation obtained in this study (24 hrs, 48 hrs, 7 days), which gives an idea of the soil moisture conditions during measurement dates, no correlation could be established between precipitation and oxidation efficiency. The bottom of the top cover at C2-N appears to have been generally prone to saturation, with accumulated water at 120 cm depth, possibly due to the capillary barrier effect (Berger et al., 2005). An exception to this was in December 2012 when conditions were relatively dry, i.e. only 4.5 mm accumulated 7 days prior to sampling and none 48 hours prior to sampling. Despite the favourable conditions on this date, i.e. no accumulation of water and no obstruction of gas flow, no oxidation was observed. Further field investigation is be needed to explain this phenomenon.

3.5 Suitability of the gas profile method

The gas profile method is subject to some limitations, particularly regarding steady state conditions and microbial soil respiration. Steady state conditions are rarely encountered in the field as gas flow rates vary continuously with precipitation and changes in atmospheric pressure. Only further investigation would permit better understanding of the influence of the lack of steady state and of microbial respiration on the results presented herein. A larger data set would also improve global Eff_{ox} estimations, as under- and overestimations due to this assumption would even each other out.

The method assumes that respiration is negligible compared to CO_2 produced due to CH_4 oxidation. In soils with little or no organic matter content, this assumption is respected. In the case of Fíflholt, with an organic matter content of the soil at around 7%, it can be assumed

that respiration plays a minor role in the total production of CO_2, accounting nonetheless for a certain error in Eff_{ox} calculations that can only be quantified through further examination. In a batch experiment using soil with total organic carbon 4.9% - 7.5%, CO_2 respiration accounted for 1.2 - 1.9% of the observed CO_2 production from CH_4 oxidation (Gebert et al., 2011b), and the oxidation efficiency was only slightly overestimated using a soil with 6% organic matter. The gas profile method would however not suit a soil with a higher organic matter content, as this would need to be accounted for in the design of a passive methane oxidation biosystem in Iceland.

4. Conclusions

Given the results obtained, the current top cover at Fíflholt can be considered as a passive methane oxidation biosystem, capable of oxidizing a significant fraction of the landfill gas passing through it, albeit not on a continuous basis. The top cover was however not designed with this purpose in mind. It was installed to comply with the operational permit, requiring a 1-m thick soil cover, which is why considerations were not made regarding the base of the cover where the layer of decomposing shredded wood may be prone to moisture retention, altering upward gas transport. Appropriate measures have to be taken in the design of a methane oxidation biocover, with particular regard to gas distribution at the base of the top cover and soil geotechnical properties.

It is of particular interest that the cover at Fíflholt was observed to oxidize methane despite generally low temperatures within the top cover, and that other factors, e.g. soil water content, may be a more controlling factor for methane oxidation. Thorough field studies are needed to provide a better understanding of the role of different soil specific and climatic factors in the oxidation process and will also prove useful in the near future for all landfills regardless of gas collection requirements.

In modelling gas emissions for National Inventory Reports on greenhouse gas emissions, a default oxidation factor of 10% is recommended in the IPCC guidelines for industrial countries with well managed landfills, although a factor of 0% has been used thus far for Iceland. The default value has however been questioned, particularly at landfills or in countries where measurements have demonstrated much higher oxidation efficiencies (e.g. Scharff & Jacobs, 2006; Chanton et al., 2009). A field study which is conducted on a larger landfill area and includes measurements during all climatic seasons would improve modelling emissions estimates and would verify the applicability of the current default oxidation factor.

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